Experimental entanglement of four particles*

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Quantum mechanics allows for many-particle wave functions that cannot be factorized into a product of single-particle wave functions, even when the constituent particles are entirely distinct. Such "entangled" states explicitly demonstrate the nonlocal character of quantum theory, have been suggested for use in high-precision spectroscopy, and are a fundamental element of schemes for quantum communication, cryptography, and computation. In general, the more particles which can be entangled, the more clearly nonclassical effects are exhibited, and the more useful the states are for quantum applications. In pursuit of these goals, we have demonstrated a recently proposed entanglement technique applicable to trapped ions. Coupling between the ions is provided by the Coulomb interaction through their collective motional degrees of freedom, but actual motional excitation is minimized. Entanglement is achieved using a single laser pulse, and the method can in principle be applied to any number of ions. We report here the use of this technique to generate entangled states of two, and for the first time, four particles.

Most experimental demonstrations of entanglement to date have relied on the selection of data from random processes, such as the preparation and detection of photon pairs in parametric down-conversion, 7-9 or of atoms in a thermal beam. 10 All methods of this type suffer from inescapable signal degradation when entanglement of larger numbers of particles is attempted, since the probability of randomly generating the appropriate conditions decreases exponentially. For instance, in the experiment of Ref. 7, two-photon entangled states could be generated and detected at a rate of roughly 1000/s, three-photon states at a rate of 30 per hour, and four-photon states at an extrapolated rate of several per year. Trapped ions have been suggested as a system in which such effects might be avoided, 11 and in a previous experiment, we demonstrated two-particle entanglement in a deterministic way. 12 By "deterministic" we mean that the desired state could be produced with a high degree of certainty at a user-specified time, 13 which is necessary for avoiding the degradation described above. However, that experiment relied on the particular behavior of two ions in a quadrupole RF trap, and could not easily be applied to larger numbers of particles.

The entanglement technique proposed by Mølmer and Sørensen^{6,14} can be understood by considering a pair of spin-1/2 charged particles confined together in a harmonic potential. The energy levels of this system are illustrated in Fig. 1, where $\hbar\omega_0$ is the internal energy

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splitting of each particle, and ν is the oscillation frequency of a particular collective mode of the particles in the trap. Using laser-cooling and optical-pumping techniques, ¹⁵ the particles are initially prepared in their spin-down internal state and in the ground state of their collective motion: $|\psi\rangle = |\downarrow\downarrow 0\rangle$. By applying optical fields oscillating at $\omega_0 + \nu - \delta$ and $\omega_0 - \nu + \delta$, the two-step transition from $|\downarrow\downarrow 0\rangle$ to $|\uparrow\uparrow 0\rangle$ is driven. For sufficiently large δ , the intermediate states $|\uparrow\downarrow 1\rangle$ and $|\downarrow\uparrow 1\rangle$ are negligibly occupied, so that no motional excitation actually occurs. The resulting interaction Hamiltonian, in the rotating-wave approximation and the Lamb-Dicke limit, is then

$$H = \frac{\hbar \tilde{\Omega}}{2} \Big(|\uparrow\uparrow\rangle\langle\downarrow\downarrow| + |\downarrow\downarrow\rangle\langle\uparrow\uparrow| + |\uparrow\downarrow\rangle\langle\downarrow\uparrow| + |\downarrow\uparrow\rangle\langle\uparrow\downarrow| \Big), \tag{1}$$

with $\tilde{\Omega} = \eta^2 \Omega^2 / \delta$ when the single particle $|\downarrow\rangle \leftrightarrow |\uparrow\rangle$ transition has Rabi frequency Ω and the Lamb-Dicke parameter is η . For an excitation involving momentum transfer $\hbar k$ and a total particle mass of M, η is given by $(\hbar k^2/2M\nu)^{1/2}$. Entanglement is achieved by applying H for a time $t = \pi/2\tilde{\Omega}$, making the spin wavefunction $|\psi_2\rangle = (|\uparrow\uparrow\rangle - i|\downarrow\downarrow\rangle)/\sqrt{2}$. This spin state is in fact created for any initial motional state $|n\rangle$, so long as the Lamb-Dicke criterion $\eta^2 n \ll 1$ is satisfied.

In order for the intermediate states $|\uparrow\downarrow 1\rangle$ and $|\downarrow\uparrow 1\rangle$ to be negligibly occupied, the detuning δ must be large compared to the transition linewidth $\eta\Omega$. However, it is clear from the expression for $\tilde{\Omega}$ that the entanglement speed is maximized for small δ , and in fact the technique can still be applied for $\delta \approx \eta\Omega$. Although motional excitation does then occur to some degree, for select values of δ the excitation vanishes at precisely the time that the entangled spin state is created. The condition for this to occur is

$$\delta/\eta\Omega = 2\sqrt{m} \tag{2}$$

for any integer m, and the maximum excitation during the pulse then has mean quantum number $\bar{n} = 1/2m$. Our experiment is operated with m = 1.

As Mølmer and Sørensen discuss, their entanglement method is scalable in the sense that precisely the same operation can be used to generate the N-particle entangled state

$$|\psi_N\rangle = (|\uparrow\uparrow\dots\uparrow\rangle + i^{N+1}|\downarrow\downarrow\dots\downarrow\rangle)/\sqrt{2}$$
 (3)

if N is any even number, while for N odd, $|\psi_N\rangle$ can be generated using one entanglement pulse accompanied by a separate independent rotation of each particle's spin.

If the ions are uniformly illuminated, the Mølmer and Sørensen scheme requires that they all participate equally in the intermediate motional excitation, which implies that the only suitable mode for arbitrary N is the center-of-mass mode. However, this mode has a practical disadvantage because in our experiments fluctuating ambient electric fields cause it to heat at a significant rate. Although for large δ the entanglement operation is largely independent of the motion, so that heating is unimportant, in the small- δ case it is necessary that motional decoherence be avoided. Modes involving only relative ion motion couple to higher moments of the field, so heating of them is negligible, ¹⁵ and for N=2 and N=4, such modes do exist in which each particle participates with equal amplitude. ¹⁷ In both cases, they are symmetric "stretch" modes, in which alternating ions oscillate out of phase. We use

these modes here. Excitation of the center-of-mass mode does still affect the experiment, as the motion in spectator modes modifies the coupling strength to the mode of interest.^{14,15} For this reason, we initially cool both the center-of-mass and stretch modes to near their ground state.

The experiment was performed using ${}^9\mathrm{Be}^+$ ions confined in a miniature linear RF trap, 18 with the N ions lying in a line along the trap's weak axis. Two spectrally resolved ground-state hyperfine levels compose the effective spin-1/2 system, with $|\downarrow\rangle \equiv |F=2,m_F=-2\rangle$, $|\uparrow\rangle \equiv |F=1,m_F=-1\rangle$, where F is the total angular momentum quantum number, and $\hbar m_F$ is the projection of the angular momentum along the quantization axis defined by an externally applied magnetic field. The hyperfine splitting $\omega_0/2\pi$ is approximately 1.25 GHz. Coherent coupling between $|\downarrow\rangle$ and $|\uparrow\rangle$ is provided via stimulated Raman transitions. The two Raman laser beams have a wavelength of 313 nm with a difference frequency near ω_0 , and are perpendicular, with their difference wave-vector lying along the line of ions. They are detuned \sim 80 GHz blue of the $2P_{1/2}$ excited state, with intensities giving $\Omega/2\pi \approx 500$ kHz. For both the two- and four-ion experiments, the desired stretch-mode frequency $\nu/2\pi$ was 8.8 MHz, giving $\eta=0.23/N^{1/2}$. The two driving frequencies required for the entanglement operation are generated by frequency modulating one of the Raman beams using an electro-optic modulator.

After the entanglement operation, the ions are probed by illuminating them with a circularly-polarized laser beam tuned to the $2S_{1/2}(F=2,m_F=-2)\leftrightarrow 2P_{3/2}(F=3,m_F=-3)$ cycling transition. Each ion in $|\downarrow\rangle$ fluoresces brightly, leading to the detection of ~ 15 photons/ion on a photomultiplier tube during a 200 μ s detection period. In contrast, an ion in $|\uparrow\rangle$ remains nearly dark. Because the number of photons detected from a spin-down ion fluctuates according to Poisson statistics, in a single experiment the the number of spin-down ions can be determined with only a limited accuracy. For the data reported, each experiment was repeated 1000 times under the same conditions, and the resulting photon-number distribution fit to a sum of Poissonians to determine the P_j . The results are given in Table I, and show that in both cases, the probabilities for all N ions to be in the same state are large compared to the probabilities for the other cases. This is characteristic of the states $|\psi_N\rangle$, although the fact that the middle probabilities are nonzero indicates that we do not generate the entangled states with perfect accuracy.

In order to prove that we are generating a reasonable approximation to $|\psi_N\rangle$, it is necessary to prove that the populations of $|\uparrow \dots \uparrow\rangle$ and $|\downarrow \dots \downarrow\rangle$ are coherent. In terms of the density matrix for the system, ρ , we must measure the far off-diagonal element $\rho_{\uparrow \dots \uparrow, \downarrow \dots \downarrow}$, whose amplitude will be abbreviated $\rho_{(\uparrow \downarrow)}$. This can be achieved by applying a simple analysis pulse to the ions before observing them. If the Raman difference frequency is set to ω_0 (and the frequency modulator turned off), each ion i undergoes ordinary Rabi oscillations, evolving according to the Hamiltonian

$$H_i = \frac{\hbar\Omega}{2} \left(e^{i\phi} |\uparrow\rangle_i \langle\downarrow|_i + e^{-i\phi} |\downarrow\rangle_i \langle\uparrow|_i \right), \tag{4}$$

where ϕ is the phase of the difference frequency relative to that of the entanglement pulse. This Hamiltonian is applied for time $\pi/2\Omega$ (a $\pi/2$ pulse), and the parity

$$\Pi \equiv \sum_{j=0}^{N} (-1)^j P_j \tag{5}$$

is observed while ϕ is varied. As seen in Fig. 2, for N ions Π oscillates as $\cos N\phi$, and the amplitude of this oscillation is in fact $2\rho_{(\uparrow\downarrow)}$. The resulting values are given in Table I. From the data shown in the table, our state preparation fidelity

$$F \equiv \langle \psi_N | \rho | \psi_N \rangle = \frac{1}{2} \left(P_{(\uparrow)} + P_{(\downarrow)} \right) + \rho_{(\uparrow\downarrow)} \tag{6}$$

can be determined, where $P_{(\uparrow)}$ is the population of $|\uparrow \dots \uparrow\rangle$ and $P_{(\downarrow)}$ is the population of $|\downarrow \dots \downarrow\rangle$. For N=2 we achieve $F=0.83\pm0.01$, while for N=4, $F=0.57\pm0.02$.

The fact that $\rho_{(\uparrow\downarrow)}$ is nonzero is still insufficient to guarantee entanglement. To be explicit, a system with density matrix ρ exhibits N-particle entanglement only if no decomposition $\rho = \sum_k p_k |\psi_k\rangle \langle \psi_k|$ exists with all the $\{|\psi_k\rangle\}$ factorizable into products of wavefunctions which depend on fewer than N particles. For example, if $|\psi\rangle_i = (|\uparrow\rangle_i + |\downarrow\rangle_i)/\sqrt{2}$ is a state of ion i, then the four-particle state $|\psi\rangle_1|\psi\rangle_2|\psi\rangle_3|\psi\rangle_4$ is not entangled, but still has $\rho_{(\uparrow\downarrow)} = 1/16$. We note that these are the types of states studied in liquid-state nuclear magnetic resonance experiments.¹⁹ Alternatively, for $|\psi\rangle_{12} = (|\uparrow\rangle_1|\uparrow\rangle_2 + |\downarrow\rangle_1|\downarrow\rangle_2)/\sqrt{2}$ and $|\psi\rangle_{34} = (|\uparrow\rangle_3|\uparrow\rangle_4 + |\downarrow\rangle_3|\downarrow\rangle_4)/\sqrt{2}$, the state $|\psi\rangle_{12}|\psi\rangle_{34}$ exhibits two-particle, but not four-particle entanglement, and has $\rho_{(\uparrow\downarrow)} = 1/4$.

To establish that we are actually observing N-particle entanglement, consider an arbitrary factorizable wave function

$$|\psi_F\rangle = [a|\uparrow...\uparrow\rangle_X + b|\downarrow...\downarrow\rangle_X + ...][c|\uparrow...\uparrow\rangle_Y + d|\downarrow...\downarrow\rangle_Y + ...], \tag{7}$$

where X and Y refer to two distinct subsets of the N particles, with $|\uparrow \dots \uparrow\rangle_X$ indicating the state with all particles in subset X spin up, and similarly for the other terms. Normalization of the factor wavefunctions requires $|a|^2 + |b|^2 \le 1$ and $|c|^2 + |d|^2 \le 1$, which can be combined and rewritten as

$$(|a| - |c|)^{2} + 2|ac| + (|b| - |d|)^{2} + 2|bd| \le 2.$$
(8)

Since the squared terms on the left are positive, (8) implies that $|ac| + |bd| \le 1$, and in turn that $(|ac| + |bd|)^2 \le 1$. Expanding the square yields the desired relation,²⁰

$$P_{(\uparrow)} + P_{(\downarrow)} + 2\rho_{(\uparrow\downarrow)} = 2F \le 1, \tag{9}$$

where $P_{(\uparrow)} = |ac|^2$, $P_{(\downarrow)} = |bd|^2$, and $\rho_{(\uparrow\downarrow)} = |abcd|$ are the previously defined quantities. Since (9) holds for any separable wavefunction, it must also hold for any separable density matrix, and is therefore true of any unentangled system. Both our N = 2 and N = 4 experiments give F > 1/2, so the states they produce exhibit N-particle entanglement.

Quantifying the amount of entanglement present is a more difficult question. A variety of measures of entanglement have been proposed, but most are difficult to calculate even numerically.^{21,22} For N=2, Wootters has given an explicit formula for the "entanglement of formation," E, as a function of ρ .²³ Although we have not reconstructed the entire two-particle density matrix, the populations measured place sufficient bounds on the unmeasured elements to determine that $E\approx 0.5$. This indicates that roughly two pairs of our ions would be required to carry the same quantum information as a single perfectly entangled pair.

In the four-ion case, no explicit formula for entanglement is known. The data do indicate that our density matrix can be expressed approximately as

$$\rho = 0.43|\psi_4\rangle\langle\psi_4| + 0.57\rho_{\text{incoh}},\tag{10}$$

where $|\psi_4\rangle$ is the desired state and ρ_{incoh} is completely incoherent (ie., diagonal). These coefficients are determined directly from the value of $\rho_{(\uparrow\downarrow)}$ in Table I, together with the fact that no evidence for other off-diagonal matrix elements was observed. To determine a measure of entanglement, however, it is necessary to decompose ρ as a sum of $|\psi_4\rangle$ and a "worst-case" factorizable matrix ρ_F , which can be accomplished as

$$\rho = 0.13|\psi_4\rangle\langle\psi_4| + 0.87\rho_F. \tag{11}$$

Note that Eqs. (10) and (11) both describe the same physical state, but that in (11), ρ_F consists of a specific mixture of two- and three-particle entangled states that is highly unlikely to occur in our experiments. In either description, it is clear that our state-preparation accuracy is limited.

The source of decoherence in our experiments is not entirely clear, but evidence suggests that it is related to intensity fluctuations in the Raman laser beams, a problem we are working to understand and correct. The presence of decoherence, and the fact that it affects the fourion experiment more strongly than the two-ion one, illustrates the need to carefully define the sense in which our entanglement operation is "scalable." Any entanglement experiment is exponentially sensitive to decoherence as the number of particles involved is increased, unless sufficient accuracy can be achieved for error-correction schemes to be gainfully applied. Such schemes are thought to require an error rate on the order of 10^{-4} per operation, and we are certainly far from this regime. However, even if such a level of fidelity were to be achieved, applications such as quantum computing still require very large entangled states to be generated in a reasonable amount of time and using a reasonable amount of resources. The method demonstrated here is important in this regard, since it uses only a single operation and requires a time that scales roughly as $N^{1/2}$.

In conclusion, we have demonstrated for the first time a four-particle entangled state, and have done so with an intrinsically scalable method. In the language of quantum information science, we have realized a four-quantum-bit logic gate. This system is relevant for the future development of quantum information technology, as such states may used to implement quantum error-detection schemes²⁶ or to demonstrate rudimentary algorithms.^{27,28} Entanglement of four particles is also interesting in its own right, as such states can show strong violations of local realism.⁵ Even the two-particle Bell's inequality measurement would be interesting to implement, as the near-perfect detection efficiency for ions would eliminate the "fair sampling" hypothesis which has been required in other experiments.²⁹ In addition to improved fidelity, applications such as these do require the ability to perform individual manipulation and detection of each ion, but this is not expected to be a severe experimental challenge.³⁰

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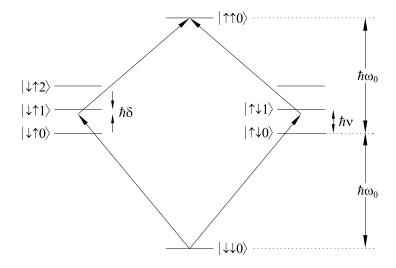
Correspondence and requests for materials should be addressed to C.M. (email: monroe@boulder.nist.gov).

						$ ho_{(\uparrow\downarrow)}$
2	0.43	0.11	0.46	-	-	0.385
4	0.35	0.10	0.10	0.10	0.35	0.215

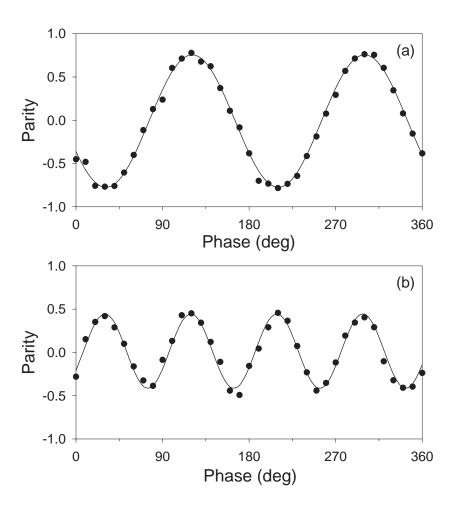
Table 1: Characterization of two-ion and four-ion states. P_j denotes the probability that j ions were measured to be in $|\downarrow\rangle$, and $\rho_{(\uparrow\downarrow)}$ denotes the amplitude of the density matrix element $\rho_{\uparrow...\uparrow,\downarrow...\downarrow}$. Uncertainties in $\rho_{(\uparrow\downarrow)}$ and the N=2 populations are ± 0.01 , and uncertainties in the N=4 populations are ± 0.02 .

Figure 1: Entanglement scheme for two particles. Each ion is initially prepared in the $|\downarrow\rangle$ internal state, and the collective motion of the pair is cooled to its ground state $|0\rangle$. Laser fields oscillating near $\omega_0 + \nu$ and $\omega_0 - \nu$ couple the $|\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\rangle$ states as shown. By detuning the single transition frequencies by a small amount δ , the populations of the $|\downarrow\uparrow1\rangle$ and $|\uparrow\downarrow1\rangle$ states are kept small. Then by driving the double transition for the appropriate time, the entangled state $(|\uparrow\uparrow\rangle - i|\downarrow\downarrow\rangle)/\sqrt{2}$ is created. For four ions, the same procedure generates the state $(|\uparrow\uparrow\uparrow\uparrow\rangle + i|\downarrow\downarrow\downarrow\downarrow\rangle)/\sqrt{2}$. Note that in the actual experiment, each of the single transitions shown is itself a two-photon Raman transition, driven by a pair of laser beams; the entire process therefore consists of a four-photon transition.

Figure 2: Determination of $\rho_{(\uparrow\downarrow)}$ for (a) two ions and (b) four ions. After the entanglement operation of Fig. 1, an analysis pulse with relative phase ϕ is applied on the single-ion $|\downarrow\rangle\leftrightarrow|\uparrow\rangle$ transition. As ϕ is varied, the parity of the N ions oscillates as $\cos N\phi$, and the amplitude of the oscillation is twice the magnitude of the density-matrix element $\rho_{(\uparrow\downarrow)}$. Each data point represents an average of 1000 experiments, corresponding to a total integration time of roughly 10 s for each graph.



Sackett et al. Figure 1



Sackett et al., Figure 2